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Energy efficiency of advanced oxidation processes for the decomposition of persistent organic compounds in water using plasmas

プラズマを用いた促進酸化による水中難分解有機物の分解エネルギー効率

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An advanced oxidation process (AOP) using OH radicals is a promising method for decomposing persistent organic substances in water. The ozone and hydrogen peroxide (H_2O_2) method is widely used to form OH radials; however, this method needs a continuous supply of O_3 and H_2O_2. We have developed a new plasma AOP system in which H_2O_2 is continuously supplied by plasmas generated in oxygen bubbles. Sample wastewater containing highly concentrated persistent substances was completely decomposed within 6 h of operation. The energy efficiency for the decomposition is a function of the production efficiency of O_3 and H_2O_2. The highest H_2O_2 yield of 2.1 g/kWh was obtained at 2.5 kV, 4 kHz, and a 1-μs pulse width.

1. Introduction

An advanced oxidation process (AOP) is one of the most effective methods for decomposing persistent organic pollutants in water using OH radicals. Typical AOP systems feed ozone gas and a hydrogen peroxide (H_2O_2) solution into wastewater. However, the continuous supply of H_2O_2 has some difficulty; thus, the in-situ generation of H_2O_2 is particularly needed for processing high concentrations and a large amount of wastewater.

This report presents a new plasma-based AOP system in which both O_3 and H_2O_2 are supplied by plasmas [1]. This system needs no supply of H_2O_2 solution and can mineralize highly concentrated persistent organics in wastewater. The generation efficiency of H_2O_2 [2]-[4] was studied to improve the system efficiency.

2. Experimental setup

Fig. 1 shows the experimental setup of the plasma-based AOP system in which O_3 and H_2O_2 are generated by an AC power supply and a capacitor discharging circuit, respectively. The capacitance of storage capacitor was 2 nF. An acrylic chamber contained 50 mL of wastewater. A ceramic having a tiny hole with a diameter of 0.3 mm separated the wastewater and the gas chamber. Oxygen flowed through the hole at a flow rate of 100 mL/min and generated gas bubbles 3–5 mm in diameter in the water. A pulsed plasma was generated inside the bubbles. H_2O_2 was generated through the interaction between the water surface and the plasma.

The H_2O_2 concentration cannot be measured in wastewater; therefore, the generation efficiency of H_2O_2 was evaluated with a 1% Na_2SO_4 solution. The H_2O_2 concentration was measured by PACKTEST Hydrogen Peroxide [Kyoritsu Chemical-Check Lab, WAK-H H_2O_2] with an absorptiometer (JASCO, V-630).

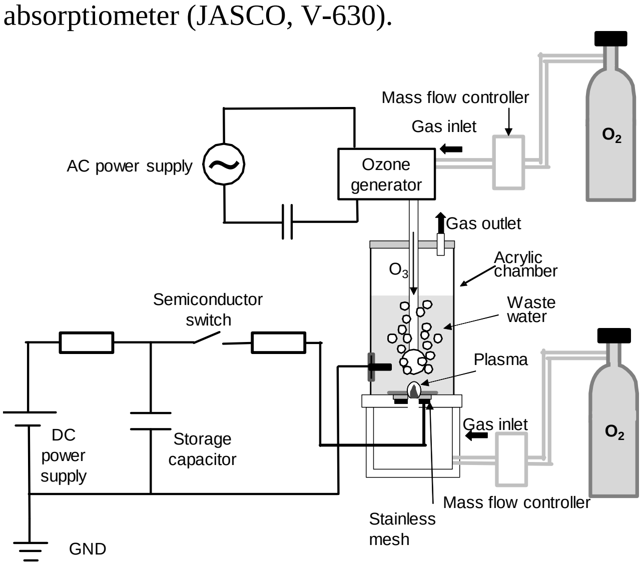

Fig. 1. Experimental setup of the plasma-based AOP system.

3. Experimental results

Fig. 2 shows the results obtained for 50 mL of wastewater treated by O_3 and H_2O_2, which was generated by the pulsed plasma. The average discharge input power was 5.7 W, and the ozone concentration was 105 g/Nm^3. After 480 min of operation, 99% of the persistent compounds in the wastewater were mineralized. The energy efficiency of the decomposition process was estimated to be 0.3 g_{TOC}/kWh. The efficiency was not so high; nonetheless, the complete decomposition of highly concentrated organic compounds in wastewater was achieved. To improve the energy efficiency of decomposition, the generation efficiencies of OH, H_2O_2, or O_3 by discharge should be improved in addition to the dissolution efficiency of ozone.

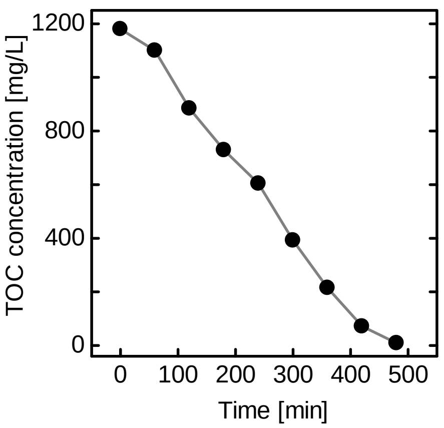

Fig. 2. Time variation of the TOC concentration of wastewater treated using the plasma and ozone system.

The generated H_2O_2 rates and yields were studied by varying the plasma parameters of repetitive frequency, applied voltage, pulse width, and the amount of water vapor in the oxygen gas. Increasing the pulse frequency could increase the generation rate while keeping the energy yield constant, although the rate of increase in the generation rate decreased at high frequencies. Increasing the applied voltage was inefficient for increasing the generation rate because the energy yield decreased. The effect of pulse width was evaluated with a constant discharge power at 1 kHz. With a larger pulse width, the generation rate and energy yield both increased as shown in Fig. 3. These results indicate that low-voltage, long-pulse-width, and high-frequency operation are suitable for the generation of H_2O_2. Introducing water into the supply gas improved the energy yield of H_2O_2 when the amount of water was greater than the amount in the saturated water vapor. A maximum generation rate of 61.8 mg/h and an energy yield of 2.1 g/kWh were obtained at 2.5 kV, 4 kHz, and a 1-μs pulse width.

The applied voltages and pulse widths were 3.5, 4, and 5 kV and 100, 200, and 300 ns, respectively. Both the generation rate and energy yield increased with increasing pulse width. The experimental results indicate that a low-voltage long-pulse-width pulsed discharge is the suitable for the efficient production of H_2O_2.

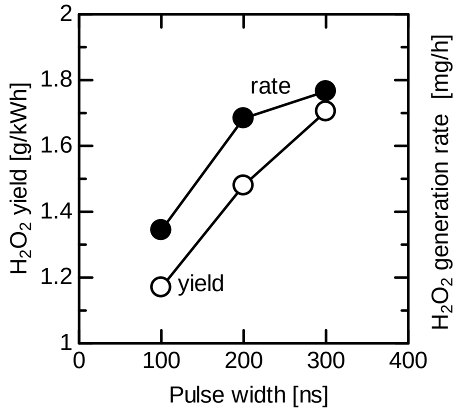

Fig. 3. H_2O_2 energy yield and generation rate as a function of the pulse width.

4. Conclusion

A plasma-based O_3 and H_2O_2 system for an AOP was developed for high-conductivity wastewater containing a high concentration of persistent compounds. A wastewater volume of 50 mL with a concentration of 1180 mg_{TOC}/L was mineralized after 480 min of operation using a pulsed plasma operated at 5.7 W in combination with 105 g/Nm^3 of ozone gas.

The properties of H_2O_2 generation by pulsed discharge were investigated by varying the plasma parameters of voltage, frequency, pulse width, and the amount of water vapor in the gas. The results indicate that low-voltage, long-pulse-width, and high-frequency operation are the most suitable for the generation of H_2O_2. A maximum generation rate of 61.8 mg/h and an energy yield of 2.1 g/kWh were obtained at 2.5 kV, 4 kHz, and a 1-μs pulse width.

Acknowledgments

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